Resonant Enhancement of Parametric Processes via Radiative Interference and Induced Coherence

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Nonlinear optical parametric processes are studied in a resonantly driven multilevel system displaying quantum interference effects. It is shown that in such systems a new regime of nonlinear amplification is possible, in which a pair of correlated Stokes and anti-Stokes fields can be generated from infinitesimally small initial values. An atomic coherence grating emerges *during* this process of efficient nonlinear amplification. The present analysis explains the results of recent optical phase conjugation experiments involving atomic phase coherence. [S0031-9007(98)07176-2]

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Since the early days of nonlinear optics there has been a substantial interest in utilizing resonant atomic and molecular systems for efficient nonlinear optical processes [1]. However, the efforts to realize the full potential of these systems have been obscured by the problems associated with resonant absorption, phase shifts, and unwanted nonlinearities leading to, e.g., self-focusing and beam distortion. The recent work, based on the concept of electromagnetically induced transparency (EIT) [2], has made encouraging progress toward solutions of these problems [3]. This work is based on the cancellation of resonant absorption and elimination of the refractive index in dark resonances or coherent population trapping [4] and has led to other exciting developments such as lasing without inversion [5].

Among the most impressive results are the experiments on nonlinear frequency conversion involving the generation of near maximal coherence using EIT [6], which follows from the earlier studies involving coherent trapping [4,7]. Here, resonant fields drive the system into the so-called "dark state," which is subsequently used as an atomic local oscillator for parametric conversion of the weaker, off-resonant fields. The use of the dark states allows one to avoid resonant absorption and phase shifts and, at the same time, to generate large nonlinearities.

The goal of the present contribution is to show that in resonant systems a large initial coherence is not always required for efficient nonlinear amplification. We demonstrate that under certain conditions interference effects can result in efficient parametric gain, and cause a pair of correlated Stokes and anti-Stokes fields as well as an atomic coherence grating to grow from small initial values, e.g., from vacuum fluctuations. Specifically, we analyze the parametric amplification in a resonant double-Λ system [8,9] [Fig. 1(a)], in which a large lower-level coherence is not established in advance. We show that in such a system large nonlinearities (on the order of "bare" resonant absorption) can coexist with suppressed absorption and phase shifts, even in the absence of the totally decoupled dark state. The analysis reveals an interesting role of quantum interference of transitions between radiatively broadened dressed states corresponding to Fig. 1(a).

We demonstrate the implications of the present mechanism of nonlinear optical enhancement by considering parametric interactions in two different propagation geometries. In the case of copropagating fields, the exponential growth of weak Stokes and anti-Stokes fields occurs, which ultimately results in the evolution of the system towards establishing a dark state and matching of the fields' amplitudes and phases. In the case of counterpropagating driving fields, a different final state is approached. Here, a substantial parametric gain and spontaneous, mirrorless oscillation can take place within a distance corresponding to few resonant absorption lengths. Possible applications of the present system include optical phase conjugation [8] and nonlinear spectroscopy [10], as well as quantum noise suppression and quantum correlation [11].

One of the key results of the present work can be understood by analyzing expressions for the polarizations $\mathcal{P}_i$ induced by the weak anti-Stokes $\mathcal{E}_1$ and Stokes $\mathcal{E}_2$

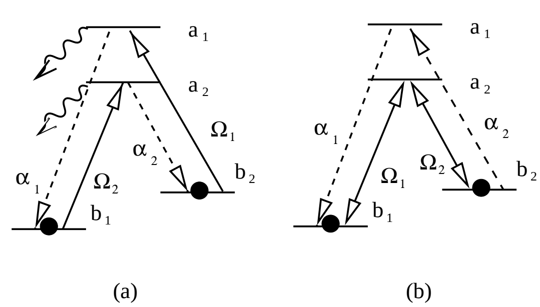

FIG. 1. (a) Parametric amplification in a generic double-Λ system. Decays are outside of the system for an open system model and into the lower metastable levels for a closed system. (b) Parametric conversion based on coherent population trapping in a double-Λ system.

fields with respective Rabi frequencies α_1, α_2 in a system [Fig. 1(a)] dressed by two strong fields E_1 and E_2 with respective Rabi frequencies Ω_1, Ω_2. In the simple case of a homogeneously broadened system with symmetric decay rates, equal drive Rabi frequencies ($|\Omega_1| = |\Omega_2| \equiv \Omega$), weak saturation of optical transitions, vanishing relaxation of the lower-level coherence ($\gamma_c \rightarrow 0$), and at the point where the detunings (Δ) of all four fields are equal, these polarizations take the form

$$\mathcal{P}_{1,2} = i(\wp_1 \wp_2 N\, \Omega_1 \Omega_2)/(2\hbar \Gamma_0 |\Omega|^2) \exp(i \delta \vec{\mathrm{k}} \cdot \vec{\mathrm{r}}) \mathcal{E}^*_{2,1} \,. \tag{1}$$

Here we have defined $\Gamma_0 = \gamma_0 + i\Delta$, where γ_0 is the decay rate of the optical coherence, $\delta\vec{\mathrm{k}} = \vec{\mathrm{k}}_{d1} + \vec{\mathrm{k}}_{d2} - \vec{\mathrm{k}}_1 - \vec{\mathrm{k}}_2$ is the geometrical phase mismatch, and k_j are the wave vectors of the drive, anti-Stokes ($j = 1$), and Stokes ($j = 2$) fields. Here and below, all polarizations, field strengths, and Rabi frequencies are slowly varying parts of the corresponding oscillating quantities.

An interesting aspect of the dressed polarizations given by Eq. (1) is that each of them (e.g., $\mathcal{P}_1$) does not depend upon the optical field driving the corresponding transition (e.g., $\mathcal{E}_1$). At the same time, the weak field acting on the adjacent transition induces a polarization which resembles the linear response of a usual two level system. Hence, according to Eq. (1), the medium of Fig. 1(a) can be transparent for each of the Stokes and anti-Stokes fields in the absence of the other. These fields, however, interact strongly via a large cross-coupling nonlinearity, resulting in an exponential growth as they propagate collinearly in the medium [Fig. 2(a)]. An even faster growth and mirrorless parametric oscillation is taking place in the case of counterpropagating fields [Fig. 2(c)].

It is instructive to compare the origin of the present result with that of usual coherent trapping in double-Λ systems [4,7] and, in particular, with maximal coherence studies of Ref. [6] [Fig. 1(b)]. Here, absorption cancellation is achieved by accumulating the population in the so-called dark superposition of the lower state energy levels. This is accomplished by applying two strong drive fields on a transition having a common upper level as in Fig. 1(b). In such a case the optical polarizations induced by the weak fields are [6,7]

$$\mathcal{P}_1 = \frac{i\wp_1}{2\hbar(\gamma - i\Delta)} \left(\wp_1 \mathcal{E}_1 - \wp_2 \frac{\Omega_2^* \Omega_1}{\Omega^2} \mathcal{E}_2 \exp(i\tilde{\mathbf{k}} \cdot \vec{\mathrm{r}}) \right), \tag{2}$$

$$\mathcal{P}_2 = \frac{i\wp_2}{2\hbar(\gamma - i\Delta)} \times \left(\wp_2 \mathcal{E}_2 - \wp_1 \frac{\Omega_2 \Omega_1^*}{\Omega^2} \mathcal{E}_1 \exp(-i\tilde{\mathbf{k}} \cdot \vec{\mathrm{r}}) \right), \tag{3}$$

where $\tilde{\mathbf{k}} = \vec{\mathrm{k}}_{d1} - \vec{\mathrm{k}}_{d2} + \vec{\mathrm{k}}_2 - \vec{\mathrm{k}}_1$, and it is assumed that $|\Omega_1| = |\Omega_2| = \Omega$ and that the weak fields are detuned equally from the respective single-photon transitions, whereas the driving fields are in exact resonance. Hence,

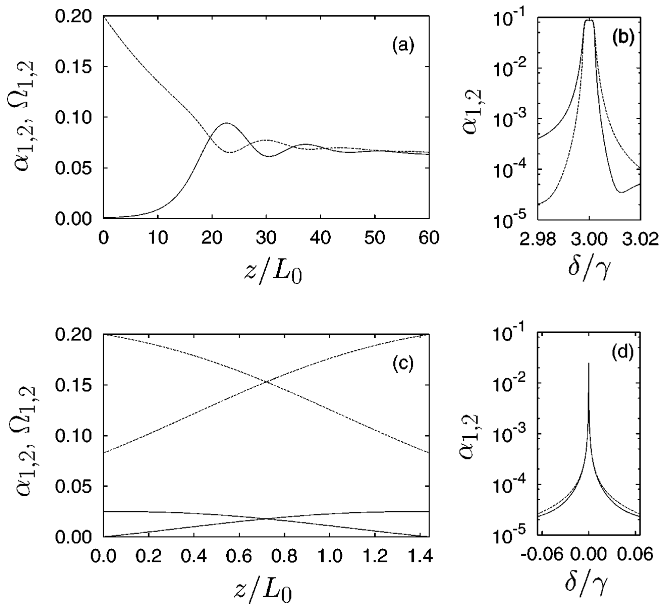

FIG. 2. Optical fields as a function of propagation distance (a),(c) and of the probe field detuning (b),(d) for copropagating geometry (a),(b) and counterpropagating geometry (c),(d). In (a) and (c) the solid curves correspond to parametrically amplified fields and the dashed curves to drive fields. In (b) and (d) the solid curves correspond to the anti-Stokes field and the dashed curves correspond to the Stokes field. The parameters are $\gamma_c = 10^{-4}\gamma$, $\Delta_{d1} = \Delta_{d2} = 3\gamma$ in (a) and (b) and $\Delta_{d1} = \Delta_{d2} = 0$ in (c) and (d). The input fields correspond to $\alpha_1^0 = 10^{-3}\gamma, 10^{-4}\gamma$ for (a) and (c), respectively, $\alpha_2^0 = 0$, and $\Omega_1^0 = \Omega_2^0 = 0.2\gamma$, where γ is the radiative decay rate on the probe transition.

we are dealing here with maximal initial coherence ($|\rho_{b1b2}| = 1/2$). Note that in this case the optical polarizations vanish when the pair of weak fields is matched such that $\Omega_2 \wp_1 \mathcal{E}_1 = \wp_2 \Omega_1 \mathcal{E}_2 \exp(i\tilde{\mathbf{k}}\vec{\mathrm{r}})$. This is, in contrast to the present result (1), indicating vanishing polarization on the probed transition (e.g., $\mathcal{P}_1 = 0$) even if $\mathcal{E}_1, \Omega_{1,2} \neq 0$ and $\mathcal{E}_2 = 0$. Furthermore, we note that the polarizations given by Eq. (1) are evaluated under the condition that the populations of the dark and the orthogonal (bright) combinations of states are approximately equal and the ground state coherence is very small ($|\rho_{b1b2}| \ll \sqrt{\rho_{b1b1}\rho_{b2b2}}$).

In order to get some insight into the physical origin of the present result, we consider in some detail the dynamics of populations and coherences in the system shown in Fig. 1. First, we note that the usual coherent population trapping occurs in the present system if only one of the driving fields, say Ω_1, is present. As the coupling of the second driving field to the medium increases (due to, e.g., increased intensity or tuning closer to the respective resonance) three principal effects take place. First of all, the second driving field can induce a nonlinearity, which can lead to the desired cross coupling between the weak optical fields. Second, the second driving field causes effective shifts of the resonance frequencies (known as light shifts). Finally, it causes population leakage out of the trapping superposition which can be envisioned as an additional relaxation process. The latter process is expected to wash out the original transparency effect. It does not happen, however, due to the following reasons. The atoms driven out of the trapping superposition do not only lead to increased absorption, but also can lead to a multiphoton amplification of the weak field. Specifically, the atom excited by, e.g., field Ω_2 into the state $|a_2\rangle$ decays spontaneously into the state $|b_2\rangle$, from which it can make a stimulated Raman-like transition back into the state $|b_1\rangle$. This process is accompanied by amplification of the weak field $\mathcal{E}_1$. As a result, the multiphoton amplification can compensate the residual absorption which preserves transparency even when the totally decoupled dark state does not exist. Such cancellation usually occurs at the point determined by the Stark shifts induced by the two driving fields. At this point the refractive indices for the two weak fields are compensated as well, in which case the polarizations are governed only by the cross-coupling nonlinearity.

We now specify the conditions, under which efficient parametric amplification is obtained. In general, the response of the dressed system to weak cw fields can be written as

$$\mathcal{P}_1 = \chi_{11}\mathcal{E}_1 + \chi_{12}\exp(i\delta\vec{k}\cdot\vec{r})\mathcal{E}_2^*, \tag{4}$$

$$\mathcal{P}_2 = \chi_{22}\mathcal{E}_2 + \chi_{21}\exp(i\delta\vec{k}\cdot\vec{r})\mathcal{E}_1^*, \tag{5}$$

where χ_{ii} are linear susceptibilities, and χ_{ij} $(i \neq j)$ describe cross coupling between Stokes and anti-Stokes components (dressed χ^3-type susceptibilities), which are responsible for parametric amplification.

The effective susceptibilities are calculated from the master equation for the density matrix describing the system of Fig. 1, and typical results are shown in Fig. 3. In general, dephasing of the coherence between the metastable levels γ_c always leads to some finite absorption at least for one of the weak fields. For example, in the case corresponding to field tunings and optical decays leading to Eq. (1), we find

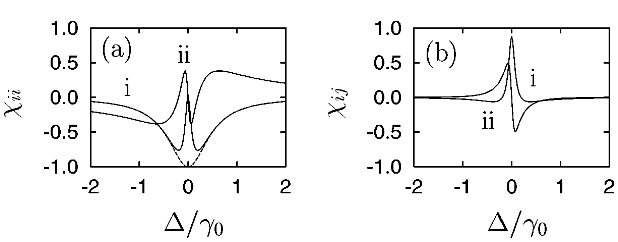

FIG. 3. Susceptibility spectra for the double-Λ parametric amplifier of Fig. 1(a). (a) Absorptive/dispersive part [$-\mathrm{Im}(\chi_{ii})$ (i) and $\mathrm{Re}(\chi_{ii})$ (ii)]. The dashed curve corresponds to $-\mathrm{Im}(\chi_{ii})$ in the case of $\Omega_1 = \Omega_2 = 0$. (b) Nonlinear cross coupling [$-\mathrm{Im}(\chi_{ij})$ (i) and $\mathrm{Re}(\chi_{ij})$ (ii)]. The drive fields are tuned exactly to the corresponding single-photon resonances. The other parameters are the same as in Fig. 2. Note that in this case $\chi_{11} = \chi_{22}$ and $\chi_{12} = \chi_{21}$.

$$\chi_{11}/\wp_1^2 = \chi_{22}/\wp_2^2 = \frac{i}{\hbar}\frac{N\Gamma_0\gamma_c(\rho_{\mathrm{uu}} - \rho_{11})}{|\Gamma_0|^2\gamma_c + 2|\Omega|^2\gamma_0}, \tag{6}$$

$$\chi_{12} = \chi_{21} = -i\frac{\wp_1\wp_2}{\hbar}\frac{N\,\Omega_1\Omega_2 2\gamma_0(\rho_{\mathrm{uu}} - \rho_{11})}{\Gamma_0(|\Gamma_0|^2\gamma_c + 2|\Omega|^2\gamma_0)}, \tag{7}$$

where $\rho_{\mathrm{uu}}(\rho_{11})$ are the populations of the upper (lower) levels, and $|\Omega_1| = |\Omega_2| = \Omega$. Clearly, in the limit $\gamma_c \to 0$ we recover Eq. (1). From the above equations, it follows that, when the Rabi frequencies of the driving fields exceed the threshold value $\Omega_{\mathrm{th}} = \sqrt{\gamma_c|\Gamma_0|}$, the nonlinearity $|\chi_{ij}|$ exceeds the linear absorption and dispersion $|\chi_{ii}|$ at the line center. Note that the threshold value of the laser intensity corresponding to Ω_{th} can be, in the case of slow lower level relaxation, much lower than the intensity required for the saturation of the optical transitions. Finally, we note that, similar to the previous studies [3,6], the large dispersion of the refractive index near resonance (Fig. 3) is important here as it can be used to eliminate any residual phase mismatch arising, e.g., due to propagation geometry, or from noncompensated light shifts.

To illustrate the significance of the present system for nonlinear parametric processes, we consider the propagation of the coupled waves in a nonlinear media of Fig. 1(a). Here, two reference waves (drive fields $E_{1,2}$) are introduced into a nonlinear medium of the length L, together with a signal wave $\mathcal{E}_1$. A new field $\mathcal{E}_2$ is generated by a four-wave mixing process. Below, we will distinguish two cases: in the first case two driving fields are assumed to propagate in the same direction, while in the second they are supposed to be counterpropagating. In each case, a weak field ($\mathcal{E}_i$) is assumed to be collinear with respective drive field (E_i). A simplified model used for the description of such a process neglects the effects of depletion and absorption of the driving fields and treats the signal and generated fields only to the first order. In the case in which all of the fields are copropagating, we find

$$\mathcal{E}_1(L) = \mathcal{E}^0\exp(\delta aL)\left[\cosh(\xi L) + \frac{a}{\xi}\sinh(\xi L)\right], \tag{8}$$

$$\mathcal{E}_2(L)^* = \mathcal{E}^0\exp(\delta aL)\frac{a_{21}}{\xi}\sinh(\xi L). \tag{9}$$

Here $a_{1j} = ik_1\chi_{1j}/2$, $a_{2j} = ik_2\chi_{2j}^*/2$, $\delta a = (a_{11} - a_{22} + i\Delta k_z)/2$, $a = (a_{11} + a_{22} + i\Delta k_z)/2$, and $\xi = \sqrt{-a_{12}a_{21} + a^2}$. Δk_z is the z component of phase mismatch $\Delta\vec{k}$, which includes the phase shifts of the driving fields. From Eqs. (8) and (9) it follows that if $\mathrm{Re}(\delta a + \xi) > 0$ both of the weak fields experience exponential growth. Note that in the absence of absorption the gain coefficient is on the order of $|k_i|\chi_{ij}$ provided that the phase matching condition is satisfied [$|k_1|\,|k_2|\mathrm{Re}\,\chi_{12}\chi_{21}^* > (\mathrm{Im}\,a)^2$], a condition which can always be adjusted for the present system by a small two-photon detuning.

The results of a numerical simulation of the complete system of equations (i.e., without small signal approximation and with propagation of all fields included) are presented in Figs. 2(a) and 2(b). Clearly, the initial period of exponential growth is followed by saturation [Fig. 2(a)]. Hence, nonlinear parametric interaction results in a matching of the fields' amplitudes and phases, after which the fields propagate as in free space. Therefore, the system evolves towards establishing a trapping state, as in matched pulse propagation [12].

A quite different evolution occurs in the case of counterpropagating fields. In this case, the solution of the linear problem yields the following expression for the generated phase-conjugate field [1]:

$$\mathcal{E}_2(0)^* = \mathcal{E}^0 \frac{1 - \exp(2\eta L)}{\exp(2\eta L)(\eta - \delta a) + (\delta a + \eta)} a_{21}, \tag{10}$$

where $\eta = \sqrt{-a_{12}a_{21} + (\delta a)^2}$. Upon inspection of Eq. (10), one finds that the amplitude of the generated field becomes large and diverges when the resonance condition $\exp(2\delta L) = (\delta a + \eta)/(\delta a - \eta)$ is fulfilled. In general, this condition gives rise to two real equations for the single variable L and can be fulfilled only under special conditions. In particular, such a solution exists when (i) δa is real, (ii) η is purely imaginary, and (iii) the length of the cell is equal to $L_r = \tan^{-1}(|\eta|/\delta a)/|\eta|$. For the present scheme at the point described by Eqs. (6) and (7) we find that if $\delta k_z = 0$ then δa is real, η is imaginary for $\Omega > \Omega_{th}$, and L_r can be as small as the resonant absorption length $L_0 = 2\hbar\gamma_0\epsilon_0/(k\wp^2 N)$. Note that δk_z can be made to vanish by properly choosing the propagation directions and two-photon detuning. That is, a large parametric gain and mirrorless oscillation is possible within a very short optical path with a drive intensity much smaller than the optical saturation intensity. These predictions were verified by numerical simulations wherein the effects of saturation and drive field propagation are included. We simulated the situation where the frequencies of all four fields are similar, for which case $\delta k_z \approx 0$. The results are shown in Figs. 2(c) and 2(d). They clearly indicate that, close to the optimal conditions described above, there is a large amplification of the signal and conjugate fields. In general, the resulting parametrically generated intensity is a substantial fraction of the initial drive intensity.

Finally, we point out that the model used in our numerical simulation describes closely the setup used in recent experiments [8] demonstrating efficient phase conjugation in a double-Λ system in sodium. The present theoretical results are in good qualitative agreement with the experiment.

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